



# Use of X-ray diffraction in assessing the aging pattern of asphalt fractions

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Received 20 February 2001; revised 1 June 2001; accepted 24 June 2001

## Abstract

The X-ray diffraction (XRD) method was employed to investigate and compare some of the crystallite parameters and aromaticity of asphaltenes obtained from fresh and aged Arabian asphalts, procured from Ras Tanura (RT) and Kuwait (KW) refineries. A few crystallite parameters in the polar aromatics, naphthene aromatics and saturate fractions of these Arabian asphalts were also determined. The results obtained by XRD method for both the types of asphaltenes and other fractions were evaluated and compared. Significant differences were observed between the structure and the aging patterns of fresh and aged asphaltenes of RT and KW asphalt. The results indicate that the source and chemistry of asphalt are responsible for the aging behavior of its components. The rolling thin-film oven and the pressurized aging vessel tests were used to simulate asphalt aging in this research. © 2001 Elsevier Science Ltd. All rights reserved.

*Keywords:* Asphaltenes; Aging; XRD

## 1. Introduction

Asphalt is a black viscous liquid obtained by refinery processes from petroleum residue. The chemical composition of asphalt is complex and varies considerably depending on the feedstock and the method of manufacture. The most widely accepted concept of the constitution is that asphalt is made up of three major components: asphaltenes, resins and oils. The consistency of asphalt can vary almost infinitely by the variations in the proportions of the chemical components.

Asphaltenes are a dark brown to black material containing large fused aromatic rings, bearing long aliphatic substituents and saturated paraffins as straight chain and branched compounds along with metals and heteroatoms as part of a ring system. The asphaltenes have been considered to be repeating units of similar composition with the major difference being in the aromaticity. The composition of maltene is very important because it contains both oils and resins which are said to be the agents that keep asphaltene molecules dispersed in the asphalt system to provide a homogeneous liquid. The aging or oxidation of the asphalt is one of the key parameters used for the characterization of asphalt properties as it directly influences the chemical composition of asphaltenes and other fractions.

X-ray diffraction (XRD) methods were developed [1] and

used by many researchers for studying and investigating the micro- and macro-structure of coal [2], carbon black [3], small aromatic system in non-crystalline polymers [4], pitch fractions [5] and oil shale kerogen [6]. In the study of asphaltenes, the XRD method was used, for the first time, by Yen et al. [7] and later several studies and meaningful results were reported [8–14].

For a discrete molecule with a simple structure, a macro-structure is sufficient to analyze and characterize the given molecule. For a much complicated and bulkier system like asphaltenes, association and micelle formation information are also needed for better characterization. The micro-structure has been defined as a short-range bonding distance between 0.5 and 2.5 Å, whereas the macro-structure relates to larger distances between 20 and 2000 Å [11]. XRD method provides an insight into the macro-structure of the asphaltenes. This method provides information about the dimension of the unit cell (crystallite parameters) such as inter-lamellar distance, layer diameter, height of the unit cell, number of the lamellar contributing to the micelle and the aromaticity [12]. These crystallite parameters also show a relationship with other physical and chemical properties of asphaltene and one of these is the molecular weight. The layer diameter ( $L_a$ ) increases with molecular weight to a certain extent; the same pattern also appears for the inter-lamellar distance ( $C/2$ ), micelle height ( $L_c$ ) and number of lamellae ( $N_c$ ) in micelles [7]. A few of these crystallite parameters are used in this study to characterize the asphaltenes isolated from the fresh and aged Arabian asphalts.

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